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(54) Title: POLYOLEFIN COMPOSITION HAVING A HIGH BALANCE OF STIFFNESS AND IMPACT STRENGTH

(57) Abstract: A polyolefin composition comprising (percentage by weight): (A) from 60 to 85% of a broad molecular weight distribution propylene polymer (component A) having a polydispersity index from 5 to 15 and melt flow rate of from 20 to 78 g/10 min (according to ASTM-D 1238, condition L); and (B) from 15 to 40% of a partially xylene-soluble olefin polymer rubber (component B) containing at least 65% by weight of ethylene. Said composition finds application in automotive field.

“POLYOLEFIN COMPOSITION HAVING A HIGH BALANCE OF STIFFNESS AND IMPACT STRENGTH”

The present invention relates to an elastomeric thermoplastic polyolefin composition. In particular, the present invention relates to compositions containing a broad molecular weight distribution propylene polymer.

Due to its mechanical and physical properties, the polymer composition of the present invention finds application above all in automotive field (e.g. bumpers and side strips).

Such a polyolefin composition has a good balance of mechanical properties, in particular improved balance of flexural modulus and IZOD impact strength even at low temperatures (e.g. at -30° C).

An added advantage, which is shown by the composition of the present invention, is that it presents low values of thermal shrinkage. Said property imparts a higher dimensional stability to the articles produced with the polyolefin composition of the present invention.

In WO00/26295 polyolefin compositions with low values of coefficient of linear thermal expansion and good mechanical properties are described, comprising (by weight) from 40 to 60% of a broad molecular weight distribution propylene polymer having a polydispersity index from 5 to 15 and melt flow rate of from 80 to 200 g/10 min (according to ASTM-D 1238, condition L); and from 40 to 60%, of a partially xylene-soluble olefin polymer rubber containing at least 65% by weight of ethylene, the IV_S/IV_A ratio between the intrinsic viscosity (IV_S) of the portion soluble in xylene of the polyolefin composition at room temperature and the intrinsic viscosity (IV_A) of the said propylene polymer ranging from 2 to 2.5.

These compositions typically have a flexural modulus of from 650 to 1000 MPa.

However, for certain automotive applications it is desirable to have compositions with flexural modulus values of higher than 1000 MPa, in particular higher than 1100 MPa, still maintaining a good balance of overall mechanical properties and low values of thermal shrinkage.

It has been surprisingly found that such balance of properties can be achieved by a combining, in compositions containing a propylene polymer and an olefin rubber, a low content of the said rubber with a particular selection of features of the propylene polymer.

Therefore an object of the present invention is a polyolefin composition comprising

(percentage by weight):

- (A) from 60 to 85%, preferably 60 to 80%, of a broad molecular weight distribution propylene polymer (component A) having a polydispersity index from 5 to 15 and melt flow rate of from 20 to 78 g/10 min, preferably from 40 to 75, more preferably from 40 to 70 g/10 min (according to ASTM-D 1238, condition L); and
- (B) from 15 to 40%, preferably 20 to 40%, of a partially xylene-soluble olefin polymer rubber (component B) containing at least 65% by weight of ethylene.

The method for measuring the xylene-soluble content and polydispersity index are described hereinbelow. The room temperature means a temperature of about 25° C in the present application.

The polyolefin composition of the present invention may further contain a mineral filler. When present, it is contained in an amount from about 0.5 to 3 parts by weight with respect to the sum of components (A) and (B).

The composition of the present invention typically has a melt flow rate of from 5 to 20 g/10 min. The intrinsic viscosity of the fraction soluble in xylene at room temperature (about 25 °C) of the overall composition is preferably of from 2 to 2.7 dl/g.

In addition, typically, it has a flexural modulus of from 1100 to 1700 MPa. Preferably the value of thermal shrinkage is from 0.5 to 1 in the longitudinal direction, and from 0.7 to 1.2 in the transversal direction; the notched IZOD resilience at -30° C is typically from 4 to 10 KJ/m². The methods for measuring the said properties are described hereinbelow.

Component (A) is a crystalline propylene homopolymer or a propylene copolymer with ethylene or C₄-C₁₀ α-olefin or a mixture thereof. Ethylene is the preferred comonomer. The comonomer content ranges preferably from 0.5 to 1.5% by weight, more preferably from 0.5 to 1% by weight.

A xylene-insoluble content at 25°C of component (A) is typically greater than 90%, preferably equal to or greater than 94%.

Component (A) preferably has a molecular weight distribution M_w/M_n , (M_w = weight average molecular weight and M_n = number average molecular weight, both measured by gel permeation chromatography) of from 8 to 30.

Preferably, component (A) comprises from 30 to 70% by weight, more preferably from 40 to 60% by weight, based on the total weight of (A), of a fraction (A¹) having melt

flow rate of from 1 to 10 g/10 min..

The olefin polymer rubber of component (B) used in the polyolefin composition of the present invention can be a poly(ethylene-*co*-C₃-C₁₀ α -olefin) or poly(ethylene-*co*-propylene-*co*-C₄-C₁₀ α -olefin) having an ethylene content preferably from 65 to 80 % by weight. The latter contains about from 0.5 to 10% by weight of a C₄-C₁₀ α -olefin. The olefin polymer rubber can optionally further contain a diene, the content of which is preferably of from 1 to 10% by weight, more preferably from 1 to 5% by weight.

The olefin polymer rubber of component (B) is partially soluble in xylene at room temperature. The xylene-insoluble content is about 25-40% by weight, preferably 30-38% by weight.

The C₃-C₁₀ α -olefins useful in the preparation of component (B) described above include propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene and 1-octene. Propylene and 1-butene are particularly preferred.

The mineral filler, when present, is preferably selected from talc, calcium carbonate, silica, conventional clays, wollastonite, diatomaceous earth, titanium oxide and zeolites. Preferably, the mineral filler is talc.

In addition to the mineral fillers discussed above, the polyolefin composition of the present invention may further contain conventional additives, for example, stabilizers, pigments, other fillers and reinforcing agents, e.g. carbon black and glass beads and fibers.

The polyolefin composition of the present invention can be prepared by way of a physical blend or chemical blend.

Preferably, the composition of the present invention is prepared directly in polymerization by sequential polymerization processes in a series of reactors based on the use of particular stereospecific Ziegler-Natta catalysts, producing by polymerization a mixture of component (A) and component (B). Subsequently, the mineral filler is, optionally, added by blending, or in the final pelletization section of the industrial polymerisation plant.

The polymerization process is carried out in at least three consecutive stages, in the presence of particular stereospecific Ziegler-Natta catalysts, supported on a magnesium halide in active form. In particular, the broad molecular weight distribution propylene polymer of component (A) described above can be prepared by sequential polymerization in at least two stages and the olefin polymer rubber in the other stage(s).

Alternatively, the polyolefin composition of the present invention can be physically blended or admixed in any conventional mixing apparatus, such as an extruder or a Banbury mixer, by mixing components (A) and (B) and optionally further components. Components (B) and (A) are blended in the molten or softened state. Thus components (A) and (B) can be prepared in separate polymerization stages, preferably with the Ziegler-Natta catalysts and the polymerization conditions hereinafter described, and then mechanically mixed.

As previously mentioned, the polymerization stage can be carried out in at least three sequential steps, wherein components (A) and (B) are prepared in separate subsequent steps, operating in each step in the presence of the polymer formed and the catalyst used in the immediately preceding step. The catalyst is added only in the first step, however its activity is such that it is still active for all the subsequent steps. The order in which components (A) and (B) are prepared is not critical. However, it is preferred to produce component (B) after producing component (A).

The catalyst used for preparing component (A) is preferably characterized in that it is capable of producing propylene polymers having a xylene insoluble fraction at 25° C greater than or equal to 90% by weight, preferably greater than or equal to 94%. Moreover, it has a sensitivity to molecular weight regulators high enough to produce propylene homopolymers having a melt flow rate in the range from 1 to 20 g/10 min and greater than 200 g/10 min.

Methods of preparing the broad molecular weight distribution propylene polymer of component (A) of the present invention are described in the European patent application 573 862.

The above said catalyst is used in all the steps of the polymerization process of the present invention for producing directly the sum of components (A) and (B).

Catalysts having the above mentioned characteristics are well known in the patent literature; particularly advantageous are the catalysts described in US patent 4,399,054 and European patents 45977 and 395083.

The polymerization process can be carried out in continuous or in batch, according to known techniques and operating in liquid phase, in the presence or absence of inert diluent, or in gas phase or in mixed liquid-gas phases. It is preferable to operate in gas phase.

Reaction time and temperature are not critical; however, the temperature typically ranges from 20 to 100° C.

Preferably, the reaction temperature is generally from 60 to 85° C for the polymerization of component (B).

Regulation of the molecular weight is carried out by using known regulators such as hydrogen.

The compositions of the present invention can also be produced by a gas-phase polymerisation process carried out in at least two interconnected polymerisation zones. The said type of process is illustrated in European patent application 782 587.

In detail, the above-mentioned process comprises feeding one or more monomer(s) to said polymerisation zones in the presence of catalyst under reaction conditions and collecting the polymer product from the said polymerisation zones. In the said process the growing polymer particles flow upward through one (first) of the said polymerisation zones (riser) under fast fluidisation conditions, leave the said riser and enter another (second) polymerisation zone (downcomer) through which they flow downward in a densified form under the action of gravity, leave the said downcomer and are reintroduced into the riser, thus establishing a circulation of polymer between the riser and the downcomer.

In the downcomer high values of density of the solid are reached, which approach the bulk density of the polymer. A positive gain in pressure can thus be obtained along the direction of flow, so that it becomes possible to reintroduce the polymer into the riser without the help of special mechanical means. In this way, a "loop" circulation is set up, which is defined by the balance of pressures between the two polymerisation zones and by the head loss introduced into the system.

Generally, the condition of fast fluidization in the riser is established by feeding a gas mixture comprising the relevant monomers to the said riser. It is preferable that the feeding of the gas mixture is effected below the point of reintroduction of the polymer into the said riser by the use, where appropriate, of gas distributor means. The velocity of transport gas into the riser is higher than the transport velocity under the operating conditions, preferably from 2 to 15 m/s.

Generally, the polymer and the gaseous mixture leaving the riser are conveyed to a solid/gas separation zone. The solid/gas separation can be effected by using conventional separation means. From the separation zone, the polymer enters the downcomer. The gaseous mixture leaving the separation zone is compressed, cooled and transferred, if appropriate

with the addition of make-up monomers and/or molecular weight regulators, to the riser. The transfer can be effected by means of a recycle line for the gaseous mixture.

The control of the polymer circulating between the two polymerisation zones can be effected by metering the amount of polymer leaving the downcomer using means suitable for controlling the flow of solids, such as mechanical valves.

The operating parameters, such as the temperature, are those that are usual in gas-phase olefin polymerisation process, for example between 50 to 120 °C.

This process can be carried out under operating pressures of between 0.5 and 10 MPa, preferably between 1.5 to 6 MPa.

Advantageously, one or more inert gases are maintained in the polymerisation zones, in such quantities that the sum of the partial pressure of the inert gases is preferably between 5 and 80% of the total pressure of the gases. The inert gas can be nitrogen or propane, for example.

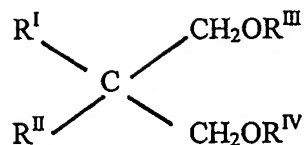
The various catalysts are fed up to the riser at any point of the said riser. However, they can also be fed at any point of the downcomer. The catalyst can be in any physical state, therefore catalysts in either solid or liquid state can be used.

An essential component of the Ziegler-Natta catalysts used in the polymerization process of the present invention is a solid catalyst component comprising a titanium compound having at least one titanium-halogen bond, and an electron-donor compound, both supported on a magnesium halide in active form.

Another essential component (co-catalyst) is an organoaluminum compound, such as an aluminum alkyl compound. An external donor is optionally added.

The solid catalyst components used in said catalysts comprise, as electron-donors (internal donors), compounds selected from the group consisting of ethers, ketones, lactones, compounds containing N, P and/or S atoms, and esters of mono- and dicarboxylic acids. Particularly suitable electron-donor compounds are phthalic acid esters, such as diisobutyl, dioctyl, diphenyl and benzylbutyl phthalate.

Other electron-donors particularly suitable are 1,3-diethers of formula:



wherein R^{I} and R^{II} are the same or different and are $\text{C}_1\text{-C}_{18}$ alkyl, $\text{C}_3\text{-C}_{18}$ cycloalkyl or $\text{C}_7\text{-C}_{18}$ aryl radicals; R^{III} and R^{IV} are the same or different and are $\text{C}_1\text{-C}_4$ alkyl radicals; or are the 1,3-diethers in which the carbon atom in position 2 belongs to a cyclic or polycyclic structure made up of 5, 6 or 7 carbon atoms and containing two or three unsaturations.

Ethers of this type are described in published European patent applications 361493 and 728769.

Representative examples of said diethers are as follows: 2-methyl-2-isopropyl-1,3-dimethoxypropane, 2,2-diisobutyl-1,3-dimethoxypropane, 2-isopropyl-2-cyclopentyl-1,3-dimethoxypropane, 2-isopropyl-2-isoamyl-1,3-dimethoxypropane and 9,9-bis(methoxymethyl) fluorene.

The preparation of the above mentioned catalyst components is carried out according to various methods. For example, a $\text{MgCl}_2 \cdot n\text{ROH}$ adduct (in particular in the form of spherical particles) wherein n is generally from 1 to 3 and ROH is ethanol, butanol or isobutanol, is reacted with an excess of TiCl_4 containing the electron-donor compound. The reaction temperature is generally from 80 to 120 °C. The solid is then isolated and reacted once more with TiCl_4 , in the presence or absence of the electron-donor compound, after which it is separated and washed with aliquots of a hydrocarbon until all chlorine ions have disappeared. In the solid catalyst component the titanium compound, expressed as Ti, is generally present in an amount from 0.5 to 10% by weight. The quantity of electron-donor compound which remains fixed on the solid catalyst component generally is 5 to 20% by moles with respect to the magnesium dihalide. The titanium compounds, which can be used for the preparation of the solid catalyst component, are the halides and the halogen alcoholates of titanium. Titanium tetrachloride is the preferred compound.

The reactions described above result in the formation of a magnesium halide in active form. Other reactions are known in the literature, which cause the formation of magnesium halide in active form starting from magnesium compounds other than halides, such as magnesium carboxylates.

The Al-alkyl compounds used as co-catalysts comprise the Al-trialkyls, such as Al-triethyl, Al-triisobutyl, Al-tri-n-butyl, and linear or cyclic Al-alkyl compounds containing two or more Al atoms bonded to each other by way of O or N atoms, or SO₄ or SO₃ groups.

The Al-alkyl compound is generally used in such a quantity that the Al/Ti ratio be from 1 to 1000.

The electron-donor compounds that can be used as external donors include aromatic acid esters such as alkyl benzoates and in particular silicon compounds containing at least one Si-OR bond, where R is a hydrocarbon radical. Useful examples of silicon compounds are (tert-butyl)₂ Si (OCH₃)₂, (cyclohexyl) (methyl) Si (OCH₃)₂, (phenyl)₂ Si (OCH₃)₂ and (cyclopentyl)₂ Si (OCH₃)₂.

1,3-diethers having the formulae described above can also be used advantageously.

If the internal donor is one of these diethers, the external donors can be omitted.

The catalysts can be precontacted with small quantities of olefins (prepolymerization), thus improving both the performance of the catalysts and the morphology of the polymers. Prepolymerization is carried out maintaining the catalysts in suspension in a hydrocarbon solvent (hexane or heptane, for example) and polymerizing at a temperature from ambient to 60° C for a time sufficient to produce quantities of polymer from 0.5 to 3 times the weight of the solid catalyst component. It can also be carried out in liquid propylene, at the temperature conditions indicated above, producing quantities of polymer that can reach up to 1000 g per g of catalyst component.

As mentioned above, the polyolefin composition of the present invention can also be obtained by blending. The blending is done using known techniques starting from pellets or powders or particles of the polymers obtained from the polymerization process, which are preferably pre-mixed with the mineral filler in the solid state (with a Banbury, Henshel or Lodige mixer, for example) and then extruded.

As above-mentioned, the polymer composition of the present invention is suitable to prepare bumpers and other parts of vehicles, such as side strips. Hence, the polymer composition is subjected to the conventional techniques used to prepare the said articles.

The following analytical methods are used to characterize the propylene polymer of component (A), rubber copolymer of component (B) and the composition obtained therefrom.

Melt Flow Rate: determined according to ASTM-D 1238, condition L.

[η] intrinsic viscosity: determined in tetrahydronaphtalene at 135° C.

Ethylene: determined according to I.R. Spectroscopy.

Soluble and insoluble in xylene: 2.5 g of polymer are dissolved in 250 ml of xylene at 135° C under agitation. After 20 minutes the solution is allowed to cool to 25° C, still under agitation, and then allowed to settle for 30 minutes. The precipitate is filtered with filter paper, the solution evaporated in nitrogen flow, and the residue dried under vacuum at 80°C until constant weight is reached. Thus one calculates the percent by weight of polymer soluble and insoluble in xylene at ambient temperature (25° C).

Polydispersity Index (P.I.): measurement of the molecular weight distribution in the polymer. To determine the P.I. value, the modulus separation at low modulus value, e.g., 500 Pa, is determined at a temperature of 200° C by using a RMS-800 parallel-plates rheometer model marketed by Rheometrics (USA), operating at an oscillation frequency which increases from 0.01 rad/second to 100 rad/second. From the modulus separation value, the P.I. value can be derived using the following equation:

$$P.I. = 54.6(\text{modulus separation})^{-1.76}$$

wherein the modulus separation (MS) is defined as:

$$MS = (\text{frequency at } G' = 500 \text{ Pa}) / (\text{frequency at } G'' = 500 \text{ Pa})$$

wherein G' is the storage modulus and G'' is the low modulus.

Flexural Modulus: determined according to ASTM-D 790.

Tensile strength at yield: ISO method 527.

Tensile strength at break: ISO method 527.

Elongation at break and at yield: ISO method 527.

Longitudinal and transversal thermal shrinkage

A plaque of 100 x 200 x 2.5 mm is moulded in an injection moulding machine "SANDRETTO serie 7 190" (where 190 stands for 190 tons of clamping force).

The injection conditions are:

melt temperature = 250°C;

mould temperature = 40°C;

injection time = 8 seconds;

holding time = 22 seconds;

screw diameter = 55 mm.

The plaque is measured 48 hours after moulding, through callipers, and the shrinkage is given by:

$$\text{Longitudinal shrinkage} = \frac{200 - \text{read_value}}{200} \times 100$$

$$\text{Transversal shrinkage} = \frac{100 - \text{read_value}}{100} \times 100$$

wherein 200 is the length (in mm) of the plaque along the flow direction, measured immediately after moulding;

100 is the length (in mm) of the plaque crosswise the flow direction, measured immediately after moulding;

the *read_value* is the plaque length in the relevant direction, measured after 48 hours.

Notched IZOD impact test at -30° C: determined according to ASTM-D 256/A.

The following examples are given in order to illustrate and not limit the present invention.

Examples 1 and 2

Preparation of the solid catalyst component

A MgCl₂/alcohol adducts in spherical form is prepared following the method described in example 2 of USP No. 2,399,054 but operating at 3,000 RPM instead of 10,000 RPM.

The adduct is partially dealcoholated by heating at increasing temperatures from 30 to 180° C operating in nitrogen current.

In a 1 liter flask equipped with a condenser and mechanical agitator is introduced, under a nitrogen current, 625 ml of TiCl₄. At 0° C while agitating are added 25 g of partially dealcoholated adduct. It is then heated up to 100° C in 1 hour; when the temperature reaches 40° C diisobutylphthalate (DIBF) is added in molar ratio Mg/DIBF=8.

The temperature is maintained at 100° C for 2 hours. It is then left to decant and afterwards the hot liquid is siphoned off. 550 ml of TiCl₄ is added and it is heated to 120° C for 1 hour. Finally, it is left to settle and the liquid is siphoned off while hot; the residual solid is washed 6 times with 200 ml aliquot of anhydrous hexane at 60° C and 3 times at room temperature. The solid is then dried under vacuum.

Polymerization

The polymerization is carried out in continuous in a series of reactors equipped with devices to transfer the product from one reactor to the one immediately next to it.

In the gas phase, hydrogen propane and monomers are continuously analyzed and fed in order to maintain constant the desired concentrations.

In the polymerization run a mixture of a triethylaluminum (TEAL) activator and dicyclopentylmethoxysilane (DCPMS) as electron-donor component is contacted with the solid catalyst component, in such a way that the TEAL/Cat weight ratio is about 5, in a reactor at 30° C for about 9 minutes. The TEAL and electron-donor compound are in such quantities that TEAL/DCPMS weight ratio is 5.

The catalyst is then transferred to a reactor containing an excess of liquid propylene and prepolymerized for 33 minutes at 25° C.

The prepolymer is then transferred to the first reactor in gas phase where the homopolymerization of the propylene occurs to obtain propylene homopolymers with low MFR. The product thus obtained is then transferred into the second reactor, where propylene is homopolymerized to obtain homopolymers with high MFR. Finally, the product of the second reactor is transferred to the third reactor, where ethylene is copolymerized with propylene to obtain component (B).

The polymerization conditions used in each reactor are shown in Table I and the properties of the products thus obtained are shown in Table II.

Table I

EXAMPLE	1	2
1° REACTOR		
Temperature (° C)	85	85
Polypropylene (wt%)	33	31.5
MFR L g/10 min	3.3	3.4
2° REACTOR		
Temperature (° C)	85	85
Polypropylene (wt%)	33	32
MFR L (g/10 min)	57.5	50.6
Xylene soluble (wt%)	2.8	2.5
IVA(dl/g)	1.00	1.00
P.I.	10.1	9.5
3° REACTOR		
Temperature (° C)	75	75
Ethylene/propylene rubber (wt%)	34	36.5
C2/(C2+C3) mol	0.481	0.503

Notes: C2 = ethylene; C3 = propylene

Table II

EXAMPLE	1	2
MFR L (g/10 min)	16.5	12.5
Xylene soluble (wt%)	24.8	26
Ethylene content (wt%)	23	25.6
IVS (dl/g)	2.34	2.46
Flexural modulus (MPa)	1264	1202
Tensile strength at yield (MPa)	22	20
Elongation at yield (%a)	7	9
Tensile strength at break (MPa)	14	15
Elongation at break (%a)	72	278
IZOD resilience at 23° C (KJ/m ²)	12.7	32
IZOD resilience at -30° C (KJ/m ²)	6.9	7.9
Longitudinal shrinkage (%)	0.73	0.90
Transversal shrinkage (%)	0.87	1.06

Note: IVS = Intrinsic Viscosity of xylene soluble fraction.

CLAIMS

1. A polyolefin composition comprising (percentage by weight):
 - (A) from 60 to 85% of a broad molecular weight distribution propylene polymer (component A) having a polydispersity index from 5 to 15 and melt flow rate of from 20 to 78 g/10 min (according to ASTM-D 1238, condition L); and
 - (B) from 15 to 40% of a partially xylene-soluble olefin polymer rubber (component B) containing at least 65% by weight of ethylene.
2. The polyolefin composition of claim 1 further containing from 0.5 to 3 parts by weight with respect to the sum of components (A) and (B) of mineral fillers.
3. The polyolefin composition of claim 1 having a melt flow rate of from 5 to 20 g/10 min.
4. The polyolefin composition of claim 1 wherein component (B) is a poly(ethylene-co-propylene).
5. A process for preparing the polyolefin composition of claim 1 where the monomers are polymerized in the presence of stereospecific catalysts supported on active magnesium halide in active form in at least three sequential steps, wherein components (A) and (B) are prepared in separate subsequent steps, operating in each step in the presence of the polymer formed and the catalyst used in the immediately preceding step.
6. Articles produced by the polyolefin composition of claim 1.
7. Bumpers produced by the polyolefin composition of claim 1.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP2004/008720

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08L23/10 B60R21/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08L B60R

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 00/26295 A (FERRARI PAOLO ; PELLICONI ANTEO (IT); GALETTI GIOVANNI (IT); MONTECH U) 11 May 2000 (2000-05-11) cited in the application the whole document	1-7
X	EP 0 928 811 A (MAZDA MOTOR) 14 July 1999 (1999-07-14)	1,2,4,6
Y	the whole document	1-7
X	EP 0 657 500 A (TOYOTA MOTOR CO LTD ; MITSUI PETROCHEMICAL IND (JP)) 14 June 1995 (1995-06-14)	1-4,6,7
Y	the whole document	1-7
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

International Application No
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